Thermal fluctuations and longitudinal relaxation of single-domain magnetic particles at elevated temperatures

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We present numerical and analytical results for the swiching times of magnetic nanoparticles with uniaxial anisotropy at elevated temperatures, including the vicinity of T_c. The consideration is based in the Landau-Lifshitz-Bloch equation that includes the relaxation of the magnetization magnitude M. The resulting switching times are shorter than those following from the naive Landau-Lifshitz equation due to (i) additional barrier lowering because of the reduction of M at the barrier and (ii) critical divergence of the damping parameters.

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The theory of thermal fluctuations of small magnetic particles is one of the fundamental issues of modern micromagnetics. The conditions at which the particle becomes superparamagnetic define the thermal stability of the magnetized state and, therefore, is also valuable for technological application such as magnetic recording [1]. The basis of the theory has been introduced by Brown [2] who suggested to include thermal fluctuations into the Landau-Lifshitz (LL) dynamical equation as formal random fields whose properties are defined by the equilibrium solution of the correspondent Fokker-Planck (FP) equation. He also derived the Arrhenius-Néel formula to describe the relaxation rate in the axially symmetric case of single-domain particles which was lately generalized to the presence of external field [3, 4]. Since the paper of Chantrell and Lyberatos [5], this Langevin-dynamics approach of Brown has been brought to numerical micromagnetics to model the thermal properties of an ensemble of interacting particles and, more generally, of ferromagnetic materials, interpreting the micromagnetic discretization elements as small particles. Generally speaking, the LL equation used in these simulations is a low-temperature approximation only. Recently a generalized Landau-Lifshitz-Bloch (LLB) equation for a ferromagnet [6, 8] was derived which is valid for all temperatures and includes the longitudinal relaxation. The deviations of the LLB dynamics from the LL dynamics should be pronounced at high temperatures, especially close to the Curie temperature T_c. The validity of this approach has been confirmed by the measurements of the domain-wall mobility in crystals of Ba-and Sr-hexaferrites close to T_c [9].

Since the proposal of the heat-assisted magnetic recording (HAMR) [10] the problem of high-temperature thermal magnetization dynamics has become of large practical importance. The basic idea of HAMR is to write bits of information at elevated temperature (close to the Curie temperature, where the switching field is small) and store the information at room temperature. To achieve a significant areal density advantage, the use of high-anisotropy intermetallics such as Ll_0 FePt has been suggested [11]. Therefore, from both fundamental and applied points of view it is necessary to consider the micromagnetics of small particles (or magnetic grains) at elevated temperatures. The straightforward approach [12] uses the formalism of the standard stochastic LL equation, however with the temperature-dependent parameters, i.e., the equilibrium magnetization $M_e(T)$ introduced through the mean-field approximation (MFA) involving the Brillouin function, and the uniaxial anisotropy $K(T)$ through the scaling relation $K(T) \sim M_e^2(T)$. However, this approach becomes invalid at elevated temperatures as it does not incorporate the longitudinal relaxational effects. The purpose of this Letter is to introduce the theoretical formalism of the thermal fluctuations of single-domain particles on the basis of the LLB equation which should be valid at all temperatures. As a practical example, we consider analytically and numerically the thermal switching of a FePt particle and discuss the conditions at which the differences between the two formalisms emerge.

We start with the LLB equation [7, 8] augmented by the white-noise fields $\boldsymbol{\zeta}$, $\boldsymbol{\zeta}_1$, and $\boldsymbol{\zeta}_2$ in the form

$$\dot{\mathbf{n}} = \gamma[\mathbf{n}\times(\mathbf{H}_{\text{eff}}+\boldsymbol{\zeta})] + \frac{\gamma\alpha_1}{n^2}(\mathbf{n}\cdot(\mathbf{H}_{\text{eff}}+\boldsymbol{\zeta}_1))\mathbf{n} - \frac{\gamma\alpha_2}{n^2}[\mathbf{n}\times[\mathbf{n}\times(\mathbf{H}_{\text{eff}}+\boldsymbol{\zeta}_2)]], \qquad (1)$$

where $\mathbf{n} \equiv \mathbf{M}/M_e(T)$, is the reduced magnetization, γ is the gyromagnetic ratio, α_1 and α_2 are dimensionless longitudinal and transverse damping parameters. The effective field $\mathbf{H}_{\text{eff}}$ is given by

$$\mathbf{H}_{\text{eff}} = -\frac{\delta F}{\delta \mathbf{M}} = \mathbf{H} + \mathbf{H}_A + (M_e/\chi_\parallel)\left(1-n^2\right)\mathbf{n}, \qquad (2)$$

where F is the free energy density of the single-domain particle (cf. Ref. [8]), $\mathbf{H}$ and $\mathbf{H}_A$ are applied and anisotropy fields, and $\chi_\parallel = \partial M_e/\partial H$ is the longitudinal susceptibility. Parameters M_e, $\chi_\parallel$, and $\alpha_{1,2}$ depend on temperature and they are singular at T_c. Within the MFA-based model, one can use Eq. (4.9) of Ref. [7] with $K_1 = K_2$ and $\gamma_{1,2} \Rightarrow \alpha_{1,2}$, rearranged to the form similar

to that of of Ref. [8]:

$$\alpha_1 = \frac{\lambda}{m_e}\frac{2T}{3T_c}\frac{2q}{\sinh(2q)}, \quad \alpha_2 = \frac{\lambda}{m_e}\left[\frac{\tanh q}{q} - \frac{T}{3T_c}\right]. \tag{3}$$

Here λ is a microscopic damping parameter that is temperature dependent but noncritical at T_c,

$$m_e \equiv M_e(T)/M_e(T=0) \tag{4}$$

is the reduced magnetization, and $q = 3T_c m_e/[2(S+1)T]$. One can see that in the vicinity of T_c the relaxational parameters diverge: $\alpha_1 \cong \alpha_2 \propto 1/M_e(T)$. In accordance with this theoretical prediction, ferromagnetic resonance measurements on permalloy have shown sharp increase of the damping close to the Curie temperature [13].

The stochastic fields in the LLB-Langevin equation above can be, in fact, introduced in many different ways. For instance, one can consider all three fields as uncorrelated, set some of them to zero, or identify some of them with each other. In all these cases one obtains different LLB-Langevin equations and different stochastic trajectories. The physical quantities obtained by averaging over realizations of $\boldsymbol{\zeta}$, $\boldsymbol{\zeta}_1$, $\boldsymbol{\zeta}_2$ are, however, the same for all models. The reason is that in all cases one obtains the same Fokker-Planck equation (FPE), as was shown in Ref. [14] for the LL-Langevin equation. The FPE corresponding to Eq. (1) can be obtained in the same way as that for the LL equation (see Appendix of Ref. [8]). The result has the form of the conservation law

$$\frac{\partial f}{\partial t} + \frac{\partial}{\partial \mathbf{n}}\cdot \mathbf{J} = 0, \tag{5}$$

where $f \equiv f(\mathbf{n},t)$ is the probability density and the probability current $\mathbf{J}$ reads

$$\begin{aligned}\mathbf{J} &= [\mathbf{n}\times\mathbf{H}_{\text{eff}}]f + \frac{\alpha_1}{n^2}\mathbf{n}\left(\mathbf{n}\cdot\left(\mathbf{H}_{\text{eff}} - \frac{T}{VM_e}\frac{\partial}{\partial\mathbf{n}}\right)\right)f \\ &\quad - \frac{\alpha_2}{n^2}\left[\mathbf{n}\times\left[\mathbf{n}\times\left(\mathbf{H}_{\text{eff}} - \frac{T}{VM_e}\frac{\partial}{\partial\mathbf{n}}\right)\right]\right]f. \end{aligned}\tag{6}$$

We will illustrate the stochastic dynamics of single-domain magnetic particles for the model with the z-uniaxial anisotropy,

$$F_A = \left(M_x^2 + M_y^2\right)/(2\chi_\perp), \tag{7}$$

where $\chi_\perp$ is the transverse susceptibility that is a constant within the MFA. We use Eq. (7) rather than $F_A = -M_z^2/(2\chi_\perp)$ to make T_c independent of the anisotropy and thus to simplify our formalism. Eq. (7) could be rewritten using a generalization of the micromagnetic anisotropy K as $F_A = \left(\nu_x^2 + \nu_y^2\right)K$ (or as $F_A = -\nu_z^2 K$), where $\boldsymbol{\nu}$ is the magnetization direction vector, $\boldsymbol{\nu} \equiv \mathbf{M}/M$ (see Ref. [8]). This is not helpful, however, within the approach based on the LLB equation. The problem is that $K = M^2/(2\chi_\perp)$ is not a constant and even not a function of temperature (cf. $K = M_e^2(T)/(2\chi_\perp)$ used in Ref. [12]), since the magnetization magnitude M can change dynamically during the thermal escape process. It is convenient to scale the free energy density as $F = \left(M_e^2/\chi_\perp\right)\tilde{F}$ with $\tilde{F}$ given by[15]

$$\tilde{F} = -\mathbf{n}\cdot\mathbf{h} + \frac{1}{2}\left(n_x^2 + n_y^2\right) + \frac{1}{4a}\left(1 - n^2\right)^2, \tag{8}$$

where $\mathbf{h} \equiv (\chi_\perp/M_e)\,\mathbf{H}$ and $a \equiv 2\chi_\|/\chi_\perp$. We also define the temperature variable σ similarly to Ref. [2]:

$$VF/T \equiv 2\sigma\tilde{F}, \qquad \sigma \equiv VM_e^2/\left(2\chi_\perp T\right). \tag{9}$$

We will restrict our consideration to the case $H = 0$. In this case the minima of $\tilde{F}$ are located at $n_x = n_y = 0$, $n_z = \pm 1$, and $\tilde{F}_{\min} = 0$. The saddle point of $\tilde{F}$ is $n_z = 0$ and $n_\perp \equiv \sqrt{n_x^2 + n_y^2} = n_s$, where

$$n_s = \begin{cases} \sqrt{1-a}, & a \le 1 \\ 0, & a \ge 1, \end{cases} \qquad \tilde{F}_{\text{sad}} = \begin{cases} (2-a)/4, & a \le 1 \\ 1/(4a), & a \ge 1. \end{cases} \tag{10}$$

In the limit $T \to 0$ (i.e., $\chi_\| \to 0$ and thus $a \to 0$) Eq. (8) confines the vector $\mathbf{n}$ to the sphere $n \equiv |\mathbf{n}| = 1$, and the standard formalism based on the LL equation is recovered. At nonzero temperatures, $a > 0$, the magnetization changes its magnitude. In our model this effect is maximal at the saddle point where the magnetization vector is perpendicular to the easy axis. One can visualize the trajectory of this vector in the process of thermal activation, after averaging over fluctuations, as an ellipsis going via the saddle point. At $T = T^*$ defined by $a = 1$ the ellipsis degenerates into a line. In the range $T^* \le T < T_c$ one has $a > 1$, and the situation is qualitatively different. Here $\mathbf{n}$ contracts preserving its direction along the z axis and goes through zero at the saddle point, then it grows in the opposite direction. These scenarios are very similar to the transformation of the domain wall structure with temperature [9]. Obviously the process of thermal activation of single-domain magnetic particles cannot be described on the basis of the LL equation at elevated temperatures. The crucial process of the longitudinal relaxation is captured by Eqs. (1)–(6) based on the LLB equation.

The escape rate in the case $T \ll \Delta U$ has the form

$$\Gamma = \Gamma_0 \exp\left(-\frac{\Delta U}{T}\right), \qquad \frac{\Delta U}{T} = \frac{VF}{T} \equiv 2\sigma\tilde{F}_{\text{sad}}. \tag{11}$$

In addition to the dependence $\sigma \propto M_e^2(T)$ in Eq. (9) that is responsible for the barrier lowering at elevated temperatures, there is another source of the barrier lowering described by $\tilde{F}_{\text{sad}}$ in Eq. (10). In particular, the value of $\tilde{F}_{\text{sad}}$ at $a = 1$ is two times smaller than its low-temperature value, $a \to 0$. The prefactor Γ_0 in Eq. (11) can be obtained by solving the FPE, Eq. (5), similarly to the derivation in Ref. [4]. For $a \lesssim 1$ the result depends on α_2 only, since in this case the barrier is being crossed by the pure rotation of the magnetization vector. For $a \gtrsim 1$ both the longitudinal and transverse relaxation becomes

important, and it is difficult to find an analytical solution to the FPE. Fortunately, in this temperature range α_1 and α_2 given by Eq. (3) already become very close to each other, so that one can set $\alpha_1 \Rightarrow \alpha_2$ everywhere. Then the calculation yields

$$\Gamma_0 = \alpha_2\omega_1\sqrt{\frac{\sigma}{\pi}}\sqrt{\frac{1-n_s^2}{a}}\exp\left[\frac{a\sigma}{2}\left(1-\frac{1}{a}\right)^2\theta(a-1)\right] \times\mathrm{erfc}\left[\sqrt{\frac{a\sigma}{2}}\left(1-\frac{1}{a}\right)\right]. \tag{12}$$

Here $\theta(x)$ is the step function and $\omega_1 = \gamma M_e/(2\chi_\perp)$ is the ferromagnetic-resonance frequency. For $a \lesssim 1$ the total rate simplifies to

$$\Gamma \cong 2\alpha_2\omega_1\sqrt{\frac{\sigma}{\pi}}\exp\left[-\sigma\left(1-\frac{a}{2}\right)\right]. \tag{13}$$

This reduces to the Brown's formula $\Gamma = 2\alpha_2\omega_1\sqrt{\sigma/\pi}e^{-\sigma}$ [2] in the limit $a \to 0$. Exactly at $a = 1$, Eqs. (11) and (12) yield $\Gamma = \alpha_2\omega_1\sqrt{\sigma/\pi}e^{-\sigma/2}$. Just below T_c according to Eqs. (8) and (9) one has $a\sigma \propto (T_c - T)^{2\beta-\gamma}$, where β and γ are the critical indices for the magnetization and susceptibility. Within the MFA $a\sigma$ remains finite at T_c, whereas for more realistic models it diverges. It makes, however, little sense to work out the appropriate limiting expressions for Γ because near T_c the high-barrier approximation $\Delta U \gg T$ becomes invalid. In fact, the prefactor Γ_0 in Eq. (11) does not strongly depend on a. The main difference of our result from the Brown's formula with a temperature-dependent barrier, $\Delta U \propto M_e^2(T)$ is described by the two factors: (i) additional lowering of the barrier because of the non-rigid magnetization, Eqs. (10) and (11); (ii) crytical divergence of the damping at T_c, Eq. (3).

Brown has obtained the $1/\sigma$ correction to the escape rate for $\sigma \gg 1$ in the form $\Gamma = 2\alpha_2\omega_1\sqrt{\sigma/\pi}e^{-\sigma}(1 - 1/\sigma)$ [16]. Within the LLB approach finding this correction in the whole range $0 \le a \le \infty$ is a complicated task. For $a \lesssim 1$ the correction factor in Eq. (12) simplifies to

$$\left[1-\frac{1}{2\sigma}\left(1+\frac{1}{n_s^2}\right)\right]. \tag{14}$$

To illustrate the practical implication of our approach, we consider thermal switching of a FePt particle (magnetic grain) at high temperature. The LLB-Langevin equation, Eq. (1) has been integrated numerically with $\boldsymbol{\zeta} = 0$, $\langle\zeta_i^\nu\rangle = 0$, and

$$\left\langle\zeta_i^\mu(t)\zeta_j^\nu(t')\right\rangle = \frac{2k_BT}{\gamma M_e\alpha_i}\delta_{ij}\delta_{\mu\nu}\delta(t-t'), \tag{15}$$

where $i,j = 1,2$ and $\mu,\nu = x,y,z$.

The microscopic relaxation parameter λ in Eq. (3) has been found analytically for a spin-phonon interaction[7]. However it is difficult to obtain reliable theoretical results for λ in general, as well as to extract λ from experiments. For our illustration below we just set $\lambda = 0.1$, neglecting its temperature dependence. The values of $m_e(T)$ of Eq. (4) can be measured or obtained from the Curie-Weiss equation $m_e = B_S(m_e\tilde{\beta})$, where $\tilde{\beta} \equiv S^2J_0/(k_BT)$, $B_S(x)$ is the Brillouin function, and J_0 is related to the experimentally measured T_c via $T_c = (1/3)S(S+1)J_0$ within the MFA. For FePt $T_c = 750$ K, and the best fit for $m_e(T)$ is obtained with $S = 3/2$ [12]. For FePt we take $M_e(T=0) = 1100$ emu/cm^3, $K(T=0) = 1.24\times10^7$ erg/cm^3, so that $\chi_\perp = M_e^2(T=0)/[2K(T=0)] = 0.0488$ Oe cm^3/emu. In the same way, the longitudinal susceptibility $\chi_{||} = \partial M/\partial H$ can be measured or found analytically from the MFA:

$$\chi_{||} = \frac{v_0M_e^2(T=0)}{S^2J_0}\frac{\tilde{\beta}B_S'(m_e\tilde{\beta})}{1-\tilde{\beta}B_S'(m_e\tilde{\beta})}, \tag{16}$$

where $v_0 = 6.4\times10^{-23}$ cm^3 is the unit-cell volume and $B_S'(x) \equiv dB_S(x)/dx$.

To integrate the LLB-Langevin equation, the Heun numerical scheme [14] has been used. The physically reasonable interpretation of the stochastical process is that in the sense of Stratonovich, as was first stressed for the LL equation in Ref. [14]. Lately, it has been shown [17] that even a naive Euler scheme which yields the Ito solution, would converge to the proper averaged physical value, if the magnetization is normalized at each time step, reflecting the conservation of the magnetization length. However, in the case of LLB equation the magnetization length is also a stochastic fluctuating variable, so that the choice of the integration scheme should explicitly include the Stratonovich interpretation.

The spins were prepared in the state $n_z = -1$, and mean first-passage time (MFPT) time was evaluated as the time elapsed until the magnetization reached the boundary value beyond the barrier, $n_z = 0.5$. The exact position of this boundary only slightly changes the MFPT. Alternatively, one can set the boundary at the top of the barrier, $n_z = 0$. In this case one has to multiply the time by 2, since in 50% of all realizations the spin crosses the barrier and in 50% of all realizations it falls back [18]. In the high-barrier case, $T \ll \Delta U$, the MFPT should coincide with the relaxation time Γ^{-1} following from the FPE.

For a comparison, we also solved the (naive) LL-Langevin equation with a constant but thermally reduced magnetization length,

$$\dot{\mathbf{n}} = \gamma[\mathbf{n}\times(\mathbf{H}_{\text{eff}}+\boldsymbol{\zeta})] - \gamma\alpha_2[\mathbf{n}\times[\mathbf{n}\times(\mathbf{H}_{\text{eff}}+\boldsymbol{\zeta}_2)]], \tag{17}$$

where $\mathbf{H}_{\text{eff}}$ is given by Eq. (2) without the last term. The temperature dependence enters this equation, as in the LLB case, via the scaling of the anisotropy energy with $M_e^2(T)$ [Eq. (7) and Eq. (8) without the last term]. The non-rigorous derivation of Eq. (17) starts with the equation $\dot{\mathbf{s}} = \gamma[\mathbf{s}\times(\mathbf{H}_{\text{eff}}+\boldsymbol{\zeta})] - \gamma\lambda[\mathbf{s}\times[\mathbf{s}\times(\mathbf{H}_{\text{eff}}+\boldsymbol{\zeta}_2)]]$ for the spin vector of unit length $\mathbf{s}$. [The same starting equation is used for the derivation of the LLB equation,

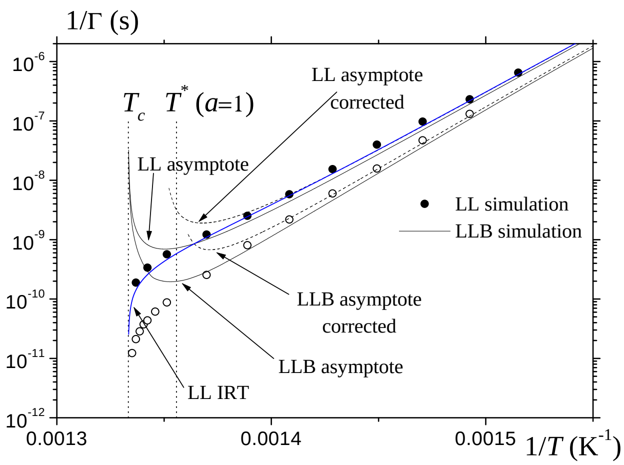

FIG. 1: Switching times for a FePt particle with 8nm diameter calculated numerically from the LL-Langevin and LLB-Langevin equations and analytically from the approproate Fokker-Planck equations. While the integral relaxation time (IRT) works very well at all temperatures, the high-barrier asymptotes break down for $\sigma \lesssim 1$. The $1/\sigma$ corrections improve the asymptotes for $\sigma \gtrsim 1$.

Eq. (1), in the classical case]. Replacing $\mathbf{s}$ in this equation by its thermal average, $\mathbf{s} \Rightarrow \mathbf{m} \equiv \langle \mathbf{s} \rangle$, and rescaling $\mathbf{m}$ as $\mathbf{m} \equiv \mathbf{n} m_e$ yields Eq. (17) with $\alpha_2 = \lambda m_e$. The latter is in a striking contradiction with the rigorous LLB expressions for the damping parameters, Eq. (3). This difference becomes very important at elevated temperatures, and there is no easy way to improve the naive LL results. In our simulations of Eq. (17) we just use the constant $\lambda = 0.1$ instead of α_2, to conform with existing publications, e.g., with Ref. [12]. Using $\alpha_2 = \lambda m_e$ leads to even more pronounced difference between the LL and LLB results.

Fig. 1 shows the MFPT of a 8 nm one-domain FePt particle as a function of temperature, calculated numerically from the LL-Langevin and LLB-Langevin equations. These numerical results are compared in Fig. 1 with Brown's analytical formula for the relaxation time Γ^{-1} [2] and the result of Eq. (12). The switching time calculated within the LLB approach is always lower than that of the LL approach due to the additional lowering of the energy barrier (10) and the critical growth of the damping at T_c. We have also shown the temperature $T^* \simeq 738$ K at which $a = 1$ and the geometry of the barrier changes. For a given particle's volume, our high-barrier approximation leading to Eq. (11) becomes invalid for $T \gtrsim T^*$. We cannot increase the volume, however, without violating the single-domain requirement.

Both Brown's formula for the LL model and Eq. (11) for the LLB model describing the Arrhenius dynamics are only valid for $T \ll \Delta U$. Switching times showing an unphysical divergence near T_c is the signature of their breakdown. For the LL model, there is a better analytical approach describing the thermally activated escape in terms of the integral relaxation time (IRT) [6, 8, 19] that is valid in the whole temperature range. The IRT for the LL model is also plotted in Fig. 1 showing a good agreement with the numerical data at all temperatures. The possibility to find the IRT analytically results from the spatial one-dimentionality of the FPE in the axially-symmetric case: $f = f(\theta, t)$. For the LLB model there are two spatial coordinates, $\{\theta, n\}$ or $\left\{ n_z, n_\perp = \sqrt{n_x^2 + n_y^2} \right\}$, and a rigorous analytical solution for the IRT seems to be impossible.

In conclusion we have introduced the formalism of the temperature fluctuations within the mean field approach based on the Landau-Lifshitz-Bloch equation. The new Langevin equation could serve as a basis for the temperature-dependent micromagnetic approach for small particles (or discretization elements) at high temperature, similar to standard temperature-dependent micromagnetics but valid in the whole temperature range. This new micromagnetics will have practical importance for the heat-assisted magnetic recording applications.

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